

PRODUCTION OF COMPOUND NUCLEI BY BOMBARDMENT OF V AND Nb WITH  
 $O^{16}$ ,  $C^{12}$ , AND  $C^{13}$  IONS

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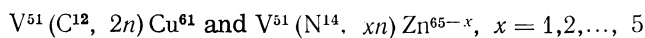
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The production of compound nuclei in the reactions  $V_{23}^{51} + O_8^{16} \rightarrow Ga_{31}^{67}$  and  $Nb_{41}^{93} + C_6^{12,13} \rightarrow Ag_{47}^{105,106}$  was investigated. The results are given in the form of excitation functions for the evaporation of different numbers of nucleons. Comparison of these reactions with reactions induced by light particles and involving compound nuclei close to  $Ga^{67}$  and  $Ag^{105}$  shows that in the case of heavy ions evaporation of a specified number of nucleons occurs at a somewhat higher excitation energy. A possible explanation is that the large angular momentum which the heavy ion contributes to the compound nucleus significantly affects the deexcitation process.

Along with the production of a compound nucleus and subsequent evaporation of nucleons, reactions were observed in which very energetic particles were emitted (60 Mev with two particles). This is not consistent with the statistical theory.

## INTRODUCTION

THE production of compound nuclei by bombarding medium-weight nuclei with multiply-charged heavy ions was studied in reference 1, where the cross sections for the reactions



were reported as functions of ion energy. In agreement with the statistical theory the excitation functions that were obtained were characteristic of neutron evaporation from uniformly heated compound nuclei. At high excitation energies, however, instead of the expected sharp falling-off of the cross section for the emission of two or three neutrons the curves were relatively flat over an interval of a few dozen Mev.

This shape of the excitation function for reactions induced by protons is accounted for by a direct interaction between the incident particle and individual nucleons of the target nucleus. Neutrons emitted as a result of such interactions possess energies considerably exceeding the energies of neutrons evaporated from compound nuclei in thermodynamic equilibrium. For reactions involving multiply-charged ions this effect was accounted for in reference 1 by the idea of "local heating," the possibility of which was pointed out by Bethe<sup>2</sup> in 1938. According to Bethe, because of the extremely short mean free path of the incident particle in nuclear matter the entire excitation energy is distributed among a few nucleons and evapora-

tion from a heated "spot" can take place before thermal equilibrium of the entire system is established. As a result of this mechanism, evaporating nucleons will obviously carry away energy much higher than that corresponding to the temperature of a uniformly heated nucleus. At high excitation energies 2 or 3 neutrons can therefore be emitted instead of 4 or 5, with correspondingly higher energies. This can account for the flat portions of the excitation functions at high energies for reactions with the emission of 2 or 3 neutrons. The detection of a similar effect when 4 or 5 neutrons are emitted would require considerably higher ion energies than those used in our experiments.

For the purpose of elucidating the reactions induced by multiply-charged ions we compared the excitation functions for these reactions with the corresponding functions for the reactions  $Cu^{63,65}(p, xn)Zn$ , where  $x = 1, 2, \dots, 5$ .<sup>3</sup> When copper is irradiated by protons the compound nuclei  $Zn^{64}$  and  $Zn^{66}$  are formed, which are close to the compound nuclei  $Cu^{63}$  and  $Zn^{65}$  that result when vanadium is bombarded by carbon and nitrogen. It was found that the excitation functions for the evaporation of a specified number of neutrons in reactions induced by multiply-charged ions are shifted by 10 to 15 Mev toward higher excitation energies. This shift may result from the fact that compound nuclei may have different momenta depending upon whether they were formed by the fusion of target nuclei with ions or with protons. For example, the compound nucleus  $Zn^{64}$  with 80

Mev excitation energy, resulting from the  $\text{Cu}^{63} + p$  reaction, has maximum angular momentum  $L_{\text{max}} \approx 10 \hbar$ ; the  $\text{Zn}^{65}$  nucleus with the same excitation energy, resulting from the  $\text{V}^{51} + \text{N}^{14}$  reaction, has  $L_{\text{max}} \approx 50 \hbar$ .

The present work has continued the study of interactions between multiply-charged ions and medium-weight nuclei, with special attention to the departures from the evaporation model. At the Atomic Energy Institute of the USSR Academy of Sciences 102-Mev oxygen ion beams were produced, making it possible to extend the investigation of interactions between multiply-charged ions and vanadium. The interactions of accelerated  $\text{C}^{12}$  and  $\text{C}^{13}$  ions with niobium were also studied.

### EXPERIMENTAL PROCEDURE

The excitation functions for the reactions  $\text{V}^{51}(\text{O}^{16}, xn)\text{Ga}$  and  $\text{Nb}^{93}(\text{C}^{12,13}, xn)\text{Ag}$  were derived by measuring the activity induced in stacked foils. In earlier experiments vanadium had been deposited on an aluminum backing, but in the present experiment the foils were made of pure vanadium and niobium.\* An activation analysis showed a lead content in the niobium of not more than 0.006%. The absence of a backing made chemical separation unnecessary and increased the likelihood of detecting products with half-lives of the order of a few minutes. This procedure also eliminates errors involved in the determination of the chemical yield and from the residual background of the backing material, and considerably reduces the error in determining the thickness of the target material.

Foils 2 to  $3 \mu$  thick were used for convenience of measurement and were irradiated in the internal beam of the 150-cm cyclotron. The energies of the  $\text{O}^{16}$ ,  $\text{C}^{13}$ , and  $\text{C}^{12}$  ions were 102, 83, and 77 Mev, respectively. In reference 4 the beam was determined to be monoenergetic to within 2%, from the line width at half maximum. The entire energy spectrum of accelerated  $\text{C}^{12}$  ions was specially studied at the cyclotron terminal radius, and it was found that low-energy ions constituted only a few per cent of the total beam intensity.

Reactions were identified mainly by means of the half-lives of  $\beta$ -active products. In some instances additional information for the identifica-

tion was obtained by determining the end-point energies of  $\beta$  spectra associated with specified half-lives; for this purpose aluminum absorbers were used.  $\beta$  particles were recorded by means of standard apparatus employing end-window counters and special holders which insured identical counting conditions for all foils and which permitted variation of the solid angle.

The apparatus used in absolute activity measurements was calibrated by means of a counter with known efficiency, which was available through the kindness of S. A. Baranov.

### INTERPRETATION OF MEASUREMENTS

Lifetimes and decay schemes in the isotope tables of reference 5 were used in calculating cross sections. This information for the reactions of interest is shown more compactly in Figs. 1 and 2, using the conventional notation. For nuclei with unknown relative  $\beta$ -decay probability in the positron-capture branch it was assumed that all decay takes place through the emission of positrons. Therefore in such cases only the lower limits of the cross sections were determined; the corresponding excitation functions are represented by dashed lines in the figures. Excitation energies were calculated using tabulated nuclear masses,<sup>6</sup> as follows:

$$E^* = (m_1 + m_2 - M)c^2 + \frac{m_2}{m_1 + m_2}E,$$

where  $m_1$ ,  $m_2$ , and  $M$  are respectively the masses of the incident ion, the target nucleus and the compound nucleus in its ground state, and  $E$  is the kinetic energy of the ion at the foil.  $E$  was determined from stopping powers given in reference 7 and the experimental range-energy curves for accelerated carbon and oxygen ions in Al, Cu, and Ni given in reference 4.

Since compound nuclei resulting from reactions with heavy ions possess considerable recoil energy, in determining the energy dependence of cross sections it was necessary to introduce a suitable correction to account for the passage of recoil nuclei from one foil to another. To this end we determined separately the mean range in vanadium of  $\text{Ga}^{67}$  nuclei produced in the  $\text{V}^{51} + \text{O}^{16}$  reaction. When 102-Mev oxygen ions are used, the energy of the  $\text{Ga}^{67}$  recoil nuclei is 24 Mev; reference 8 gives for fission fragments in this energy region the following approximate expression for the range-energy relation:

$$R = R_0(E/E_0)^{1/2}.$$

When  $R_0$  is determined experimentally at  $E_0$

\*Pure vanadium which could be rolled was provided by M. S. Makunin, a graduate student at the Metallurgical Institute, U.S.S.R. Academy of Sciences. In rolling the vanadium and niobium we were greatly assisted by B. V. Blinov of the All-Union Institute of Measurements, and by V. M. Plotko of the Joint Institute for Nuclear Research.

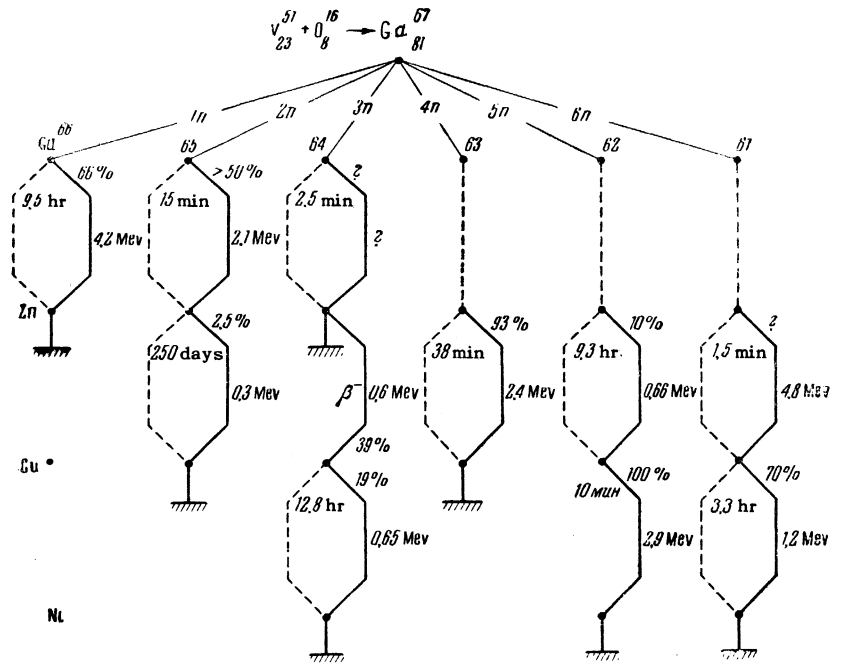


FIG. 1. Scheme of nucleon-emission and radioactive-product decay for the reaction  $V^{51} + O^{16}$ .

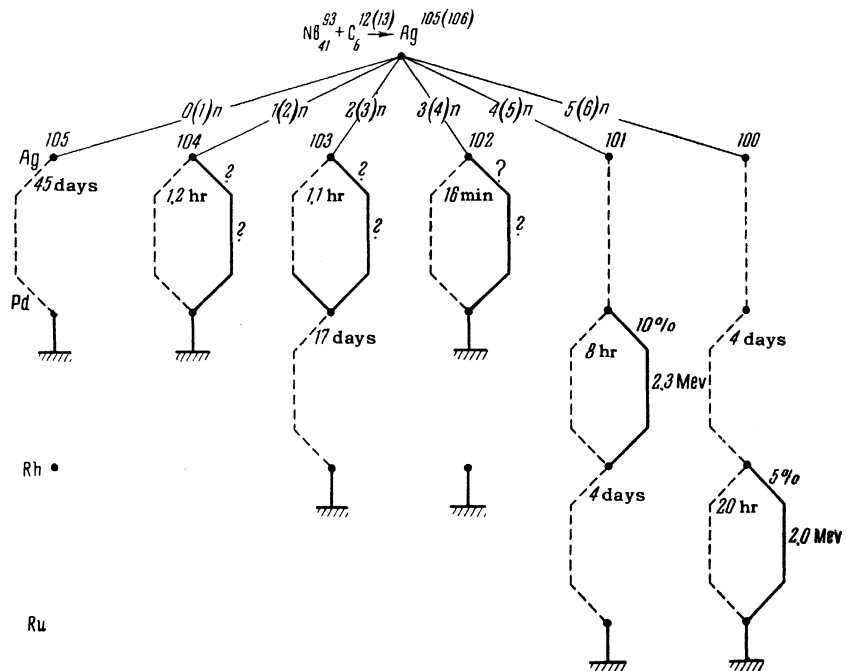


FIG. 2. Scheme of nucleon-emission and radioactive-product decay for the reaction  $Nb^{93} + C^{12,13}$ .

= 24 Mev, this formula enables us to calculate R for other energies. The calculation showed that the correction to the excitation functions to account for compound-nucleus recoil is almost independent of energy; a common shift is found for all excitation functions toward higher energies, amounting to 4 and 1 Mev for vanadium and niobium, respectively.

**ESTIMATE OF ERROR**

The sources of systematic errors in our experiments may have been errors associated with the

measurements of ion currents, inaccurate branching ratios in the tables, calibration of the measuring equipment by means of a  $4\pi$  counter and additional unaccounted-for systematic errors. When we exclude the last possibility the total systematic error of our cross section values should not exceed 17%.

Accidental errors which can distort excitation functions are much more important; these may arise through inaccurate analysis of the decay curves (5%), inaccurate weighing of the foils (1%) and inexact correspondence of foil positions

with respect to the counters (2%). Our estimates thus show that the relative cross sections were determined to within 8%.

The initial ion energies given in reference 4 were determined to within 2%; this degree of error thus applies to the energy at the first foil. As the ions are slowed down the error increases and amounts to 10% at the last foil.

## RESULTS

Figures 3–5 show the excitation functions of various reactions resulting from the irradiation of vanadium by oxygen and of niobium by carbon. The numbers of protons and of neutrons emitted by the compound nuclei are indicated without representing the order of their emission. The dashed lines pertain to reactions for which only the lower limits of the cross sections were determined. We shall now consider the curves for vanadium and niobium in greater detail.

**Vanadium.** The complete fusion of  $O^{16}$  and  $V^{51}$  results in compound nuclei of  $Ga^{67}$  with excitation energies from 39 to 90 Mev. The lower limit of the excitation energy represents kinetic energy of the oxygen ions which equals the Coulomb threshold of the reaction; the upper limit is the maximum ion energy, which was 102 Mev in our experiments.

Figure 3 shows that in the given range of excitation energies the compound nucleus decays through the emission of both neutrons and protons, with the number of evaporated particles varying from 2 to 5. The probability for the evaporation of a single neutron is evidently very small for excitation energies exceeding 40 Mev. We were unable to de-

tect a reaction product accompanied by the emission of only a single neutron, and the cross section for such a reaction must therefore be considerably smaller than 1 mb.

The  $\beta^+$  activity of  $Ga^{65}$  was used to record the reaction ( $O^{16}, 2n$ ); in this case we know only that the positrons represent more than half of the total number of decays. In the calculation of the cross section it was assumed that  $Ga^{65}$  emits only positrons; therefore the true cross section lies somewhere below twice the given value. The variation of this cross section was studied up to excitation energies of 60 Mev; at higher energies uncertainty results from the decay of  $Cu^{62}$ , which is produced in the reaction ( $O^{16}, 2p3n$ ) and has a half-life close to that of  $Ga^{65}$ .

The ( $O^{16}, 3n$ ) reaction was identified in a somewhat unusual manner. Reaction products in almost all foils of the pile exhibited  $\beta$  activity with a half-life of 2.8 hours and an end-point energy of 0.6 Mev. The shape of the excitation function and the position of its peak indicate that this activity can only result from the emission of 3 or 4 particles. The reactions ( $O^{16}, 3p$ ) and ( $O^{16}, p2n$ ) result in stable nuclei, and the ( $O^{16}, 2pn$ ) reaction is easily identified from the 12.8-hour half-life of  $Cu^{64}$ . All possible reactions involving the emission of four particles are excluded similarly; this activity was therefore assigned to  $Ga^{64}$ , which is the reaction product that accompanies three neutrons.

This identification is possible if we assume either that a) the tabular value of the  $Ga^{64}$  half-life is incorrect or b)  $Ga^{64}$  had an isomeric state

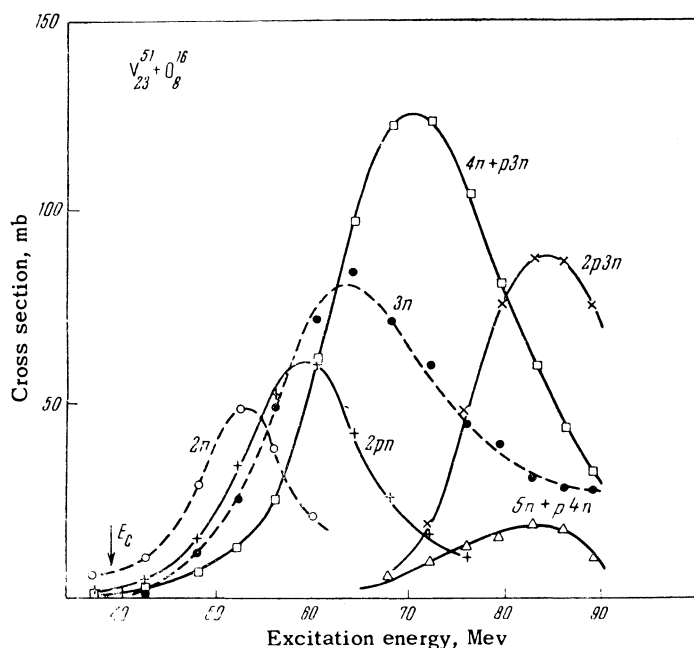


FIG. 3. Excitation functions for the reaction  $V^{51} + O^{16}$ .

with a half-life of 2.8 hours. The slow falling-off of the excitation function above 75 Mev may possibly result from the fact that at such energies the reactions  $(O^{16}, 6n)$  and  $(O^{16}, p5n)$  can occur, resulting in the formation of  $Cu^{61}$ , which has a similar half-life of 3.3 hours and 1.2-Mev end-point energy. Such activity corresponding to an excitation energy of 75 Mev was actually detected among the foils.  $Cu^{64}$  as the product of  $(O^{16}, 2pn)$  is easily detected from its 13-hour  $\beta$  activity with the spectral limit  $E_{\beta} \sim 0.5$  Mev. The excitation functions of  $(O^{16}, 4n)$  and  $(O^{16}, p3n)$  are combined since both reactions are identified by the same product  $Zn^{63}$ , with a half-life of 38 minutes and 2.4-Mev  $\beta$  end-point energy.  $Ga^{63}$ , the product of a "4n" reaction, is unknown but apparently has a short half-life.

The situation is similar with respect to the reactions  $(O^{16}, 5n)$  and  $(O^{16}, p4n)$ , which are recorded by means of the 9.3-hour emission from  $Zn^{62}$ . In this case a complication results from the small difference between the half-lives of  $Zn^{62}$  and of  $Cu^{64}$ , which is formed in a  $2pn$  reaction. The separation of the products according to  $\beta$ -particle energies is not especially accurate.

In the 75 – 90 Mev range of excitation energies 10-minute  $\beta$  activity was observed, with a peak which indicates that it must be associated with the emission of 5 or 6 particles. Figure 1 shows that this activity can only result from  $(O^{16}, 2p3n)$ , which produces  $Cu^{62}$ .

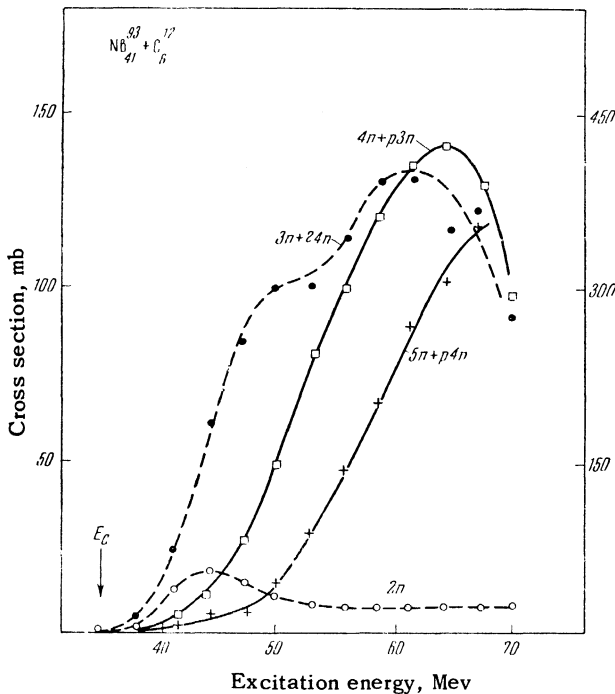


FIG. 4. Excitation functions for the  $Nb^{93} + C^{12}$  reaction. On the left – the scale for "2n" and "3n + p3n" reactions; on the right – the scale for "4n + p3n" and "5n + p4n" reactions.

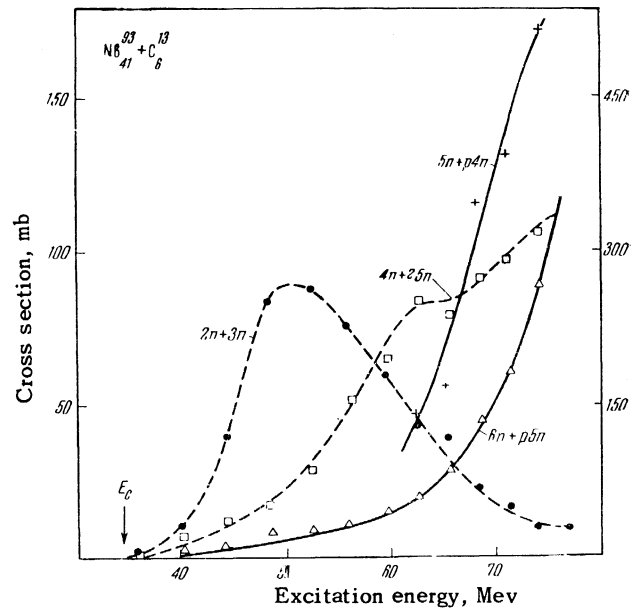


FIG. 5. Excitation functions for the  $Nb^{93} + C^{13}$  reaction. On the left – the scale for "2n + 3n" and "4n + p5n" reactions; on the right – the scale for "5n + p4n" and "6n + p5n" reactions.

**Niobium.** Excitation functions for the interaction between carbon and niobium were obtained in the excitation energy range 35 – 70 Mev. The conditions which determine the limits of this range have been considered above. The excitation functions of  $Nb^{93}(C^{12}, xn)Ag^{105-x}$  and of  $Nb^{93}(C^{13}, xn)Ag^{106-x}$  are given in Figs. 4 and 5, respectively. The method of calculating the respective cross sections in these figures will be considered together, but all quantities pertaining to the irradiation of niobium by  $C^{13}$  (Fig. 5) will be enclosed in parentheses.  $Ag^{104}$  and  $Ag^{103}$ , which are produced by the evaporation of 1 (2) and 2 (3) neutrons from the compound nucleus, have almost indistinguishable half-lives of 1.2 and 1.1 hours, respectively. We have no data on the  $\beta$ -ray end-point energies and branching of the decay schemes. Therefore the excitation function in Figs. 4 and 5 is designated by  $1n + 2n$  ( $2n + 3n$ ) and is actually the lower limit of the sum of the cross sections for the emission of 1 and 2 (2 and 3) neutrons. The evaporation of 3 (4) neutrons yields  $Ag^{102}$ , for which we also know only its 16-minute half-life. The cross section has an inflection point at 50 (65)-Mev excitation energy, very great width and a peak which is distant from the peak of the excitation function for the emission of 2 (3) neutrons. The curve appears from its shape to be the sum of two curves with a relative displacement along the horizontal axis. Some nucleus with a half-life of  $\sim 16$  minutes is evidently formed in addition to  $Ag^{102}$ ; this may be  $Ag^{101}$ , resulting from the evaporation of 4 (5) neutrons, or  $Tc^{101}$ , which results from the emission of 4 protons (4 protons + 1 neutron). The

tables contain no data on  $\text{Ag}^{101}$ ; we know that  $\text{Tc}^{101}$  exhibits  $\beta^-$  decay with a 15-minute half-life and 1.2-Mev end-point energy. We were unable to determine which of these reactions actually occurs; the lower limit of the excitation function in the figures is therefore designated arbitrarily by  $3n + 4n$  ( $4n + 5n$ ).  $\text{Ag}^{101}$ , which is formed following the evaporation of 4 (5) neutrons, decays with an unknown but apparently short half-life and is transformed into  $\text{Pd}^{101}$ , which has a half-life of 8 hours, a 2.3-Mev  $\beta^-$ -energy limit and 10% relative probability for positron emission. Aside from the reaction with the evaporation of 4 (5) neutrons,  $\text{Pd}^{101}$  may result from the emission of 3 neutrons and 1 proton (4 neutrons and 1 proton); the corresponding curves in the figure are designated by  $4n + p3n$  ( $5n + p4n$ ).

The evaporation of 5 (6) neutrons results in the production of  $\text{Ag}^{100}$ , which apparently has a very short lifetime.  $\text{Pd}^{100}$  decays purely by electron capture with a half-life of 4 days, and its daughter element, 20-hour  $\text{Rh}^{100}$ , emits positrons with energies up to 2 Mev in 5% of its decays. The identification of the reaction accompanied by the emission of 5 (6) neutrons was made by means of the 4-day activity and end-point energy of the  $\text{Pd}^{100}$   $\beta$  spectrum. However  $\text{Rh}^{101}$ , which is the product of a reaction involving the emission of 4 (5) particles, also exhibits a 4-day half-life and conversion electrons; therefore in calculating the cross section for the 5 (6) -particle case we introduced a correction determined by means of the absorption curves. The approximate character of this correction considerably reduces the accuracy of the 5 (6) -particle excitation function.

We note that when Nb was irradiated by  $\text{C}^{13}$  nuclei almost the same amount of 16-hour activity was detected in all foils. This activity, which had a cross section  $\geq 10$  mb, was not identified.

## CONCLUSIONS

From Figs. 3, 4, and 5 and from a comparison of our results with those in references 3 and 9, which present data for reactions induced by protons and  $\alpha$  particles, we may draw the following conclusions:

1. Differences in the excitation functions distinguish the competing processes in which different numbers of nucleons are evaporated from heated compound nuclei. The absolute cross sections which take into account reactions resulting in stable nuclei are so large that we may consider the formation of a stable nucleus to be the principal process for medium-weight nuclei in our range of excitation energies.

2. A comparison of the excitation function peaks observed for reactions induced by multiply-charged ions, protons and  $\alpha$  particles shows that in the case of the multiply-charged ions all excitation functions are shifted toward higher energies (by 10–15 Mev for  $\text{V} + \text{O}$  and by 7–8 Mev for  $\text{Nb} + \text{C}$ ). The probable explanation of this shift is that in all instances compound nuclei are formed with identical charges, masses and excitation energies but quite different angular momenta. The large angular momentum contributed by a multiply-charged ion creates an extra centrifugal barrier and thus results in the emission of particles with kinetic energy greater than  $2T$ ,  $T$  being the temperature of the nucleus. Moreover, the angular momentum carried away by evaporating nucleons is small, so that after the emission of a certain number of particles the residual nucleus, with an excitation energy above the nucleon binding energy, possesses such large angular momentum that particle evaporation is impossible. This residual nucleus will be deexcited through the emission of a  $\gamma$ -ray cascade. Therefore in reactions involving multiply-charged ions the energy carried away by photons is greater than the 8 Mev which is ordinarily assumed for other types of reactions.

3. To verify the departure from predictions based on the evaporation model which was observed in reference 1 (see the Introduction) the following experiments were performed: a) The  $\text{V}^{51}(\text{C}^{12}, 2n)\text{Cu}^{61}$  cross section was measured again very carefully, using pure vanadium foils and high-energy carbon ions; b) the dependence of  $\text{Nb}^{93}(\text{C}^{12}, 2n)\text{Ag}^{103}$  on ion energy was measured several times. All of these experiments showed that reactions involving the emission of two neutrons have a cross section  $\sim 10$  mb at excitation energies up to 70–80 Mev. The emission of two neutrons at such high excitation energies apparently indicates a direct interaction between the incident nucleus and nucleons of the target nucleus.

We note in conclusion that the determination of the relative probabilities of direct interactions and the production of compound nuclei will require further more detailed investigations of both the excitation functions and the angular and energy distributions of the emitted particles.

The authors are deeply indebted to Professor G. N. Flerov for valuable suggestions during the experimental work and for a discussion of the results.

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